

# Specific heat and thermal conductivity of ferromagnetic magnons in Yttrium Iron Garnet

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The specific heat and thermal conductivity of the insulating ferrimagnet  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (Yttrium Iron Garnet, YIG) single crystal were measured down to 50 mK. The ferromagnetic magnon specific heat  $C_m$  shows a characteristic  $T^{1.5}$  dependence down to 0.77 K. Below 0.77 K, a downward deviation is observed, which is attributed to the magnetic dipole-dipole interaction with typical magnitude of  $10^{-4}$  eV. The ferromagnetic magnon thermal conductivity  $\kappa_m$  does not show the characteristic  $T^2$  dependence below 0.8 K. To fit the  $\kappa_m$  data, both magnetic defect scattering effect and dipole-dipole interaction are taken into account. These results complete our understanding of the thermodynamic and thermal transport properties of the low-lying ferromagnetic magnons.

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## I. INTRODUCTION

Recently, the ferrimagnetic insulator  $\text{Y}_3\text{Fe}_5\text{O}_{12}$  (Yttrium Iron Garnet, YIG) draws great attention due to the long-range transport ability of spin current.<sup>1,2</sup> In these experiments, electronic signal can be transferred by spin angular momentum of the spin waves in insulating YIG, via the spin Hall and inverse spin Hall effects, which provides a new method to transfer information by pure spin waves.<sup>1,2</sup> In this context, it is important to understand the thermodynamic and transport properties of the spin waves.

In spin wave theory for magnets, the quanta of spin waves are magnons. There are antiferromagnetic (AFM) and ferromagnetic (FM) magnons, which have totally different dispersion relations, and distinct thermodynamic and transport properties. The AFM magnons have linear dispersion relation, so both their specific heat and boundary-limited thermal conductivity at low temperature obey the  $T^3$  dependence.<sup>3,4</sup> For FM magnons, the situation is more complex due to the existence of magnetic dipole-dipole interaction (MDDI).<sup>5,6</sup>

At not very low temperature, the dispersion relation  $E = Dk^2$  is a good approximation, and the specific heat and boundary-limited thermal conductivity of FM magnons show the characteristic  $T^{1.5}$  and  $T^2$  dependence, respectively.<sup>7-9</sup> The equations are

$$C_m(T) = \frac{15\zeta(5/2)k_B^{2.5}T^{1.5}}{32\pi^{1.5}D^{1.5}} \quad (1)$$

and

$$\kappa_m(T) = \frac{\zeta(3)k_B^3LT^2}{\pi^2\hbar D}, \quad (2)$$

where  $\zeta$  is the Riemann function,  $L$  is the boundary-limited mean free path.<sup>3,7-9</sup> However, at sub-Kelvin temperature range, the MDDI

with typical order of  $10^{-4}$  eV, has to be considered for FM magnons.<sup>10</sup> It significantly modifies the dispersion relation of FM magnons below 1 K, and makes the approximate form  $E = Dk^2$  no longer valid.<sup>11</sup>

The MDDI is long range and anisotropic. It is a basic interaction in magnets and critical for many phenomena such as the demagnetization factors and the formation of domain walls in ferromagnets, the spin ice behavior in Ising pyrochlore magnets, and the spin anisotropy in ferromagnetic films.<sup>12-14</sup> Theoretically, when the effect of MDDI was taken into account for FM magnons, both the  $T^{1.5}$  dependence of  $C_m$  and  $T^2$  dependence of  $\kappa_m$  changed.<sup>5,6,10,15</sup> Yet so far experimental verification of this effect on FM magnons is still lacking.

YIG is an archetypical ferrimagnetic insulator. Its low-energy magnetic excitations are FM magnons because at low temperature the important spin-wave branch in a ferrite has the same form as in a ferromagnet.<sup>16,17</sup> The study of this interesting and complex compound started from late 1950s, and it has become indispensable for investigating the properties of magnets since then.<sup>10</sup> In fact, YIG is the first material in which thermal conductivity contributed by magnetic excitations was observed.<sup>18</sup> At low temperature, its magnon specific heat and thermal conductivity can even exceed those contributed by phonons.<sup>18-20</sup> Therefore, YIG is an ideal compound to study the thermodynamic and transport properties of FM magnons.

Previous lowest temperature for specific heat measurement on YIG was 1 K.<sup>21,22</sup> The data of magnon specific heat  $C_m(T)$  between 1 and 4 K showed the characteristic  $T^{1.5}$  dependence.<sup>21,22</sup> The thermal conductivity of YIG was roughly measured down to 0.23 K.<sup>20</sup> The data of magnon thermal conductivity  $\kappa_m(T)$  between 0.23 and 1 K did not obey the characteristic  $T^2$  dependence, and it was explained by considering the effect of magnetic defect scattering.<sup>20,23,24</sup>

In this paper, we present the specific heat and thermal conductivity measurements of YIG single crystal down to 50 mK. The magnon specific heat data deviate down-

$$H_{d-d} = 2\mu_B^2 \sum_{i \neq j} \frac{r_{ij}^2 (\mathbf{S}_i \mathbf{S}_j) - (\mathbf{r}_j \mathbf{S}_i)(\mathbf{r}_{ij} \mathbf{S}_j)}{r_{ij}^5},$$

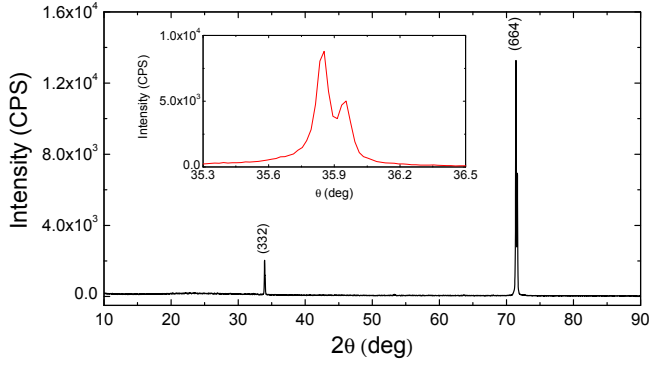


FIG. 1: (Color online) X-ray diffraction pattern for the (332) plane of YIG single crystal. Inset: rocking curve of the (664) reflection. The two peaks are from the Cu  $K_{\alpha 1}$  and  $K_{\alpha 2}$  radiations, respectively

ward from the  $T^{1.5}$  dependence below 0.77 K, which is attributed to the MDDI effect. Below 0.2 K, the magnon thermal conductivity data cannot be fitted by only considering the boundary and magnetic defect scatterings, and the MDDI effect has to be taken into account. To our knowledge, this is the first experimental observation of MDDI effect on FM magnon specific heat and thermal conductivity, giving complete understanding of the thermodynamic and thermal transport properties of the low-lying FM magnons.

## II. EXPERIMENT

The YIG single crystal was grown by the optical floating zone furnace.<sup>25–29</sup> The single crystal grew along the [332] crystallographic direction, characterized by the X-ray diffraction. Ultra-low temperature specific heat measurement was carried out on a sample with mass 45.55 mg in a small dilution refrigerator adapted into a Physical Property Measurement System (PPMS, Quantum Design company).

Sample 1 (S1) was cut from the single crystal for thermal conductivity measurements. It is rectangle shaped with dimensions  $2.08 \times 0.84 \text{ mm}^2$  in the plane and 0.63 mm thick along the [332] direction (the sample growth direction). Sample 2 (S2) was obtained by thinning S1 to 0.23 mm. For both samples, the heat current was along the  $[11\bar{3}]$  direction.

Ultra-low temperature thermal conductivity was measured in a dilution refrigerator (Oxford Instruments), using a standard four-wire steady-state method with two  $\text{RuO}_2$  chip thermometers, calibrated *in situ* against a reference  $\text{RuO}_2$  thermometer. Four contacts were made on the sample surface by silver epoxy. Magnetic fields were applied parallel to the heat current.

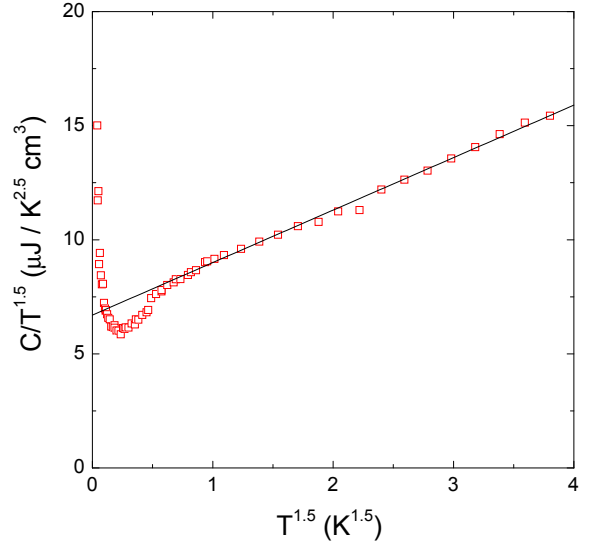


FIG. 2: (Color online) Specific heat of YIG single crystal. The data between 0.77 and 2.5 K can be fitted by the solid line  $C = 6.7T^{1.5} + 2.3T^3$ . Below 0.77 K the curve deviates from the solid line, which is attributed to the effect of magnetic dipole-dipole interaction. Note that the upturn below 0.38 K is the Schottky anomaly from nuclear moments.

## III. RESULTS AND DISCUSSION

The quality of our YIG single crystal was characterized by X-ray diffraction (XRD), as shown in Fig. 1. The main panel is the XRD pattern of the (332) plane. The inset shows the rocking scan curve of the (664) reflection, with two peaks from the Cu  $K_{\alpha 1}$  and  $K_{\alpha 2}$  radiations, respectively. The full width at half maximum (FWHM) of the peak from Cu  $K_{\alpha 1}$  is only  $0.07^\circ$ , indicating high quality of the crystal.

The specific heat of YIG single crystal below 2.5 K is shown in Fig. 2. In the figure,  $C/T^{1.5}$  was plotted as a function of  $T^{1.5}$  in order to separate phonon and magnon contributions. Between 0.77 and 2.5 K, the total specific heat can be fitted by  $C = aT^{1.5} + bT^3$ , in which the first term is from the FM magnons and the second term is from the phonons. From the fitted coefficient  $a = 6.7 \text{ μJ/cm}^3 \text{ K}^{2.5}$ , we get  $D = 5.2 \times 10^{-36} \text{ J cm}^2$  according to Eq. (1). This value is very close to Edmonds and Petersen's result  $D = 5.1 \times 10^{-36} \text{ J cm}^2$ .<sup>21</sup>

Below 0.77 K, however, there is an apparent deviation from the solid fitting line. As we have described, the MDDI will affect the specific heat of FM magnons below 1 K, by modifying the dispersion relation. The modified dispersion relation was proposed as the following form<sup>11</sup>

$$E(k) = \sqrt{(Dk^2 - N_z \hbar \omega_m)(Dk^2 - N_z \hbar \omega_m + \hbar \omega_m \sin^2 \theta_k)} \quad (3)$$

where  $\theta_k$  is the angle between the magnon wave vector and the magnetization direction,  $N_z$  is the  $z$  demagnetization factor, and  $\hbar \omega_m = g\mu_B 4\pi M$ . The dispersion relation of Eq. (3) is plotted in Fig. 3. The approxi-

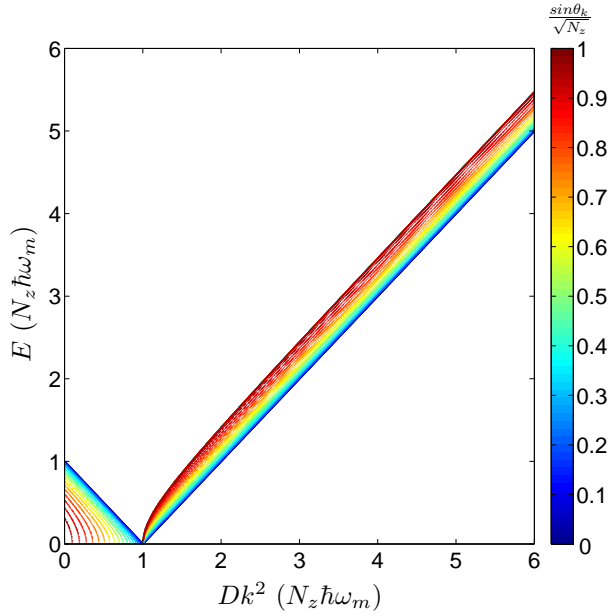


FIG. 3: (Color online) The dispersion relation of FM magnons at low energy when considering the magnetic dipole-dipole interaction effect:<sup>5</sup>  $E(k) = \sqrt{(Dk^2 - N_z \hbar \omega_m)(Dk^2 - N_z \hbar \omega_m + \hbar \omega_m \sin^2 \theta_k)}$ . It strongly deviates from the  $E = Dk^2$  approximation below the energy  $\hbar \omega_m = g\mu_B 4\pi M$ . For YIG,  $\hbar \omega_m = 0.32k_B$ .

mate dispersion relation  $E = Dk^2$  is only valid for  $k_B T \gg \hbar \omega_m$ . For YIG,  $4\pi M = 2449$  Gs,<sup>30,31</sup>  $g = 2$ ,<sup>32</sup> so  $\hbar \omega_m = g\mu_B 4\pi M = 0.32k_B$ . Therefore, the  $T^{1.5}$  dependence of  $C_m$  should only hold for  $T \gg 0.32$  K. From our experimental data in Fig. 2,  $C_m$  shows  $T^{1.5}$  dependence above 0.77 K. The downward deviation below 0.77 K should come from the effect of MDDI. Note that the upturn below 0.38 K is the Schottky anomaly from nuclear moments.

Next we discuss the thermal conductivity results. Figure 4(a) shows the ultra-low temperature thermal conductivity of YIG in magnetic fields up to 11 T, plotted as  $\kappa/T$  vs  $T$ . One can see that  $\kappa$  is strongly suppressed by field, as previously reported.<sup>18–20</sup> In the insulating YIG, the total thermal conductivity can be expressed as  $\kappa = \kappa_{ph} + \kappa_m$ , in which  $\kappa_{ph}$  and  $\kappa_m$  are the phonon and magnon thermal conductivity, respectively. Since  $\kappa_{ph}$  is usually not affected by magnetic field, the rapid suppression of  $\kappa$  with field in Fig. 4(a) should come from the reduction of  $\kappa_m$ . As we know, the external magnetic field  $H$  opens a gap  $\Delta = g\mu_B H$  in the magnon spectrum. When external field is high enough to satisfy  $g\mu_B H \gg \kappa_B T$ , there would be no magnon contribution. The saturated thermal conductivity from  $H = 4$  to 11 T shown in Fig. 4(a) suggests that there is only phonon contribution left below 0.8 K in  $H \geq 4$  T, consistent with previous reports.<sup>19,20</sup>

Therefore the FM magnon thermal conductivity  $\kappa_m$  in zero field can be extracted by subtracting  $\kappa(4\text{T})$  from  $\kappa(0\text{T})$ . In Fig. 4(b),  $\kappa_m$  is plotted as  $\kappa_m/T$  vs  $T$ . For

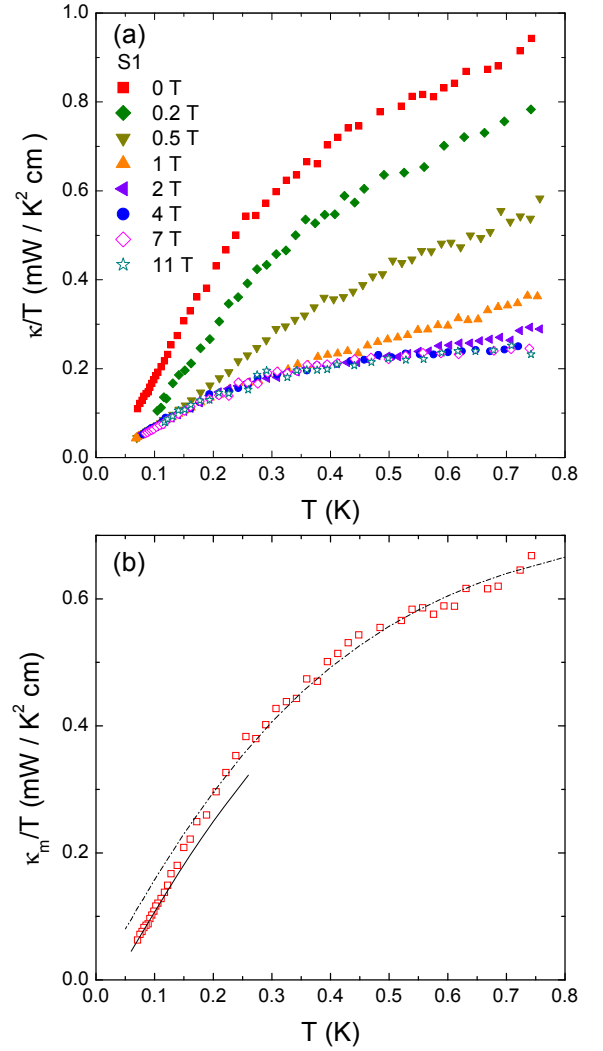


FIG. 4: (Color online) (a) Thermal conductivity of YIG single crystal in magnetic fields up to 11 T. In  $H \geq 4$  T,  $\kappa$  tends to saturate. (b) Zero field thermal conductivity of FM magnons obtained by subtracting  $\kappa(4\text{T})$  from  $\kappa(0\text{T})$ . The dashed line is the fitting of the data between 0.2 and 0.8 K, by considering the boundary scattering and magnetic defect scattering. The deviation below 0.2 K is attributed to the MDDI effect. The solid is the fitting of the data below 0.12 K by including the MDDI effect.

AFM magnons, ballistic boundary-limited  $\kappa_m = aT^3$  was observed below 0.5 K.<sup>4</sup> Apparently, for FM magnons in Fig. 4(b), there is no such a simple power-law temperature dependence.

Theoretically, the thermal conductivity of magnons is calculated by the equation

$$\kappa = \frac{k_B}{24\pi^3 \hbar} \int \left( \frac{E}{k_B T} \right)^2 \frac{e^{E/k_B T}}{(e^{E/k_B T} - 1)^2} (\nabla_k E) \ell d^3 k, \quad (4)$$

where  $\ell$  is the mean free path of magnons. At not very low temperature, if the approximate dispersion relation  $E = Dk^2$  of FM magnons is taken and only boundary scattering is considered, we get the  $T^2$  dependence of  $\kappa_m$

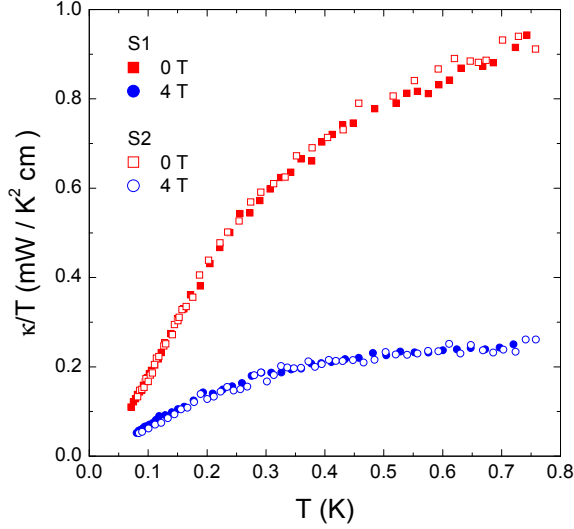


FIG. 5: (Color online) Thermal conductivity of YIG samples S1 and S2 in zero field and  $H = 4$  T. S2 is obtained by thinning S1 from 0.63 to 0.23 mm.

in Eq. (2). Therefore, the anomalous temperature dependence of  $\kappa_m$  in Fig. 4(b) must come from the MDDI effect or some other scattering mechanism. Previously, additional magnetic defect scattering was considered, and the data of  $\kappa_m$  between 0.23 and 1 K can be well fitted.<sup>20</sup> In this case,  $\ell^{-1}$  can be expressed as the sum of two terms

$$\ell^{-1} = L^{-1} + \ell_D^{-1} \quad (5)$$

where  $L^{-1}$  is from boundary scattering and  $\ell_D^{-1}$  is from magnetic defect scattering with  $\ell_D^{-1} = \alpha k^4$ . In this way, the magnon thermal conductivity is

$$\kappa_m(T) = BT^2 \int_0^\infty \frac{x^3 \text{csch}^2(\frac{1}{2}x) dx}{1 + \beta T^2 x^2}, \quad (6)$$

where  $x = E/k_B T$ ,  $B = \frac{\zeta(3)k_B^3 L}{\pi^2 \hbar D}$ ,  $\beta = \frac{\alpha L k_D^2}{D^2}$ . By using this model, our  $\kappa_m$  data between 0.2 and 0.8 K can also be well fitted, with the parameters  $B = 0.056$  mW/K<sup>3</sup> cm and  $\beta = 0.15$  K<sup>-2</sup>. Using the obtained value of  $B$ , together with  $D = 5.2 \times 10^{-36}$  J cm<sup>2</sup>, we get the boundary-limited mean free path  $L = 32.7$   $\mu$ m, which we will discuss later.

However, the magnetic defect scattering model can not fit the  $\kappa_m$  data below 0.2 K, as seen in Fig. 4(b). We further consider the effect of MDDI. Since the dispersion relation Eq. (3) is too complex to do calculation, Ortenberger and Sparks chose a simplified dispersion relation<sup>5</sup>

$$E = Dk^2 + ck_B. \quad (7)$$

Substituting this dispersion relation into Eq. (4) results in

$$\kappa_m = BT \int_0^\infty \frac{x^2 \text{csch}^2(\frac{1}{2}x)(Tx + c)}{1 + \beta(Tx + c)^2} dx. \quad (8)$$

With the  $B$  and  $\beta$  value obtained above, the  $\kappa_m$  data can be fitted well below 0.12 K, as shown in Fig. 4(b). We want to emphasize that we tried to fit the  $\kappa_m$  data below 0.8 K with only boundary scattering and MDDI effect, without considering the magnetic defect scattering, but it did not work. Therefore, the magnetic defect scattering mechanism is necessary for explain the low-temperature  $\kappa_m$  data in YIG. Comparing the results of  $C_m$  and  $\kappa_m$ , the MDDI starts to affect thermal conductivity at a lower temperature than specific heat. The reason may relate to the involvement of magnetic defect scattering in thermal conductivity.

Finally, we discuss the boundary-limited mean free path  $L$  of FM magnons in YIG. From Fig. 4(b),  $L = 32.7$   $\mu$ m is obtained by the fitting. This value is one order smaller than the expected  $L = 2\sqrt{A/\pi} = 727$   $\mu$ m for sample S1, with  $A$  being the cross section area.<sup>33</sup> Such a phenomenon has been observed previously.<sup>18,19,34</sup> Friedberg and Harris speculated that it is due to the inner boundary of thin layers rich in Fe<sup>2+</sup> inside the sample.<sup>34</sup> To test this idea, we did the same thermal conductivity measurements on sample S2, obtained by thinning S1 from 0.63 to 0.23 mm. In Fig. 5, the 0 and 4 T data of S2 are almost identical to those of S1, indicating the same  $\kappa_m$  in S1 and S2. This result shows that  $\kappa_m$  indeed does not change with the sample boundary, therefore the actual boundaries are inside the sample, likely the thin layers proposed by Friedberg and Harris.<sup>34</sup> These thin layers may form during the growing process.

#### IV. SUMMARY

In summary, by extending the measurements of the specific heat and thermal conductivity down to 50 mK, we investigate the thermodynamic and transport properties of the low-lying ferromagnetic magnons in YIG single crystal. The deviation of magnon specific heat  $C_m(T)$  from the characteristic  $T^{1.5}$  dependence below 0.77 K is attributed to the effect of magnetic dipole-dipole interaction. The magnon thermal conductivity  $\kappa_m(T)$  is extracted by subtracting  $\kappa_m(4\text{T})$  from  $\kappa_m(0\text{T})$ . Below 0.8 K,  $\kappa_m(T)$  does not obey the characteristic  $T^2$  dependence due to the magnetic defect scattering. With further decreasing temperature, the magnetic dipole-dipole interaction also shows effect on  $\kappa_m(T)$  below 0.2 K. Our work provides a complete understanding of the thermodynamic and transport properties of the low-lying ferromagnetic magnons.

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